

200504089
iAP20 Rec'd PCT/TO 25 APR 2006

DISSEMINATION APPARATUS

This invention relates to apparatus for the disseminating of volatile liquids into an atmosphere.

- 5 One very common method apparatus for disseminating a volatile liquid, such as a fragrance or an insecticide, into an atmosphere consists of a porous transfer member, such as a porous wick, that is in contact with a reservoir of volatile liquid. Liquid rises up this wick and evaporates into the atmosphere. This system has drawbacks, such as the low surface area for evaporation and the tendency for the wick to fractionate complex mixtures, such as fragrances, so that some
- 10 components are disseminated earlier than others and the full effect of the fragrance is lost.

- It has been proposed to overcome this disadvantage by using external capillaries, that is, capillary channels cut or moulded into a suitable substrate. One example is described in United States Patent 4,913,350, in which an external capillary channel-containing member is inserted
- 15 into a liquid. In another embodiment, described in United Kingdom Patent Application GB 0306449, there is fitted to a known transfer member a capillary sheet, that is, a sheet extending essentially perpendicularly from the transfer member and comprising channels of capillary dimensions, to which volatile liquid can pass and travel along for evaporation. This sheet generally contacts the transfer member by means of a hole in the sheet through which the
- 20 transfer member protrudes and within which it fits snugly, at least some of these channels contacting the transfer member such that liquid can transfer from the member to the sheet ("liquid transfer contact").

- Although this technology offers significant advantages over the porous wicks of the art, these
- 25 advantages have never been completely realized. It has now been found that it is possible to obtain the full benefits of the technology by adherence to certain fundamental parameters. The invention therefore provides an apparatus adapted to disseminate volatile liquid into an atmosphere from a reservoir, the transfer to atmosphere being at least partially achieved by means of a transfer member having external capillary channels, characterised in that

30

- (a) at least 30% by weight of the materials comprising the volatile liquid have a molecular weight of 175 maximum and the volatile liquid has a surface tension of less than 40 dynes/cm; and

BEST AVAILABLE COPY

(b) The transfer member is of plastics material having a surface energy of less than 45 dyne/cm.

By "at least 30% by weight" is meant all the components of the liquid, including any solvent present.

When the active is a fragrance it can be composed with one or more compounds, for example, natural products such as extracts, essential oils, absolutes, resinoids, resins, concretes etc., but also synthetic materials such as hydrocarbons, alcohols, aldehydes, ketones, ethers, acids, esters, acetals, ketals, nitrites, etc., including saturated and unsaturated compounds, aliphatic, carbocyclic, and heterocyclic compounds. The molecular weights range from around 90 to 320. Such fragrance materials are mentioned, for example, in S. Arctander, Perfume and Flavor Chemicals (Montclair, NJ., 1969), in S. Arctander, perfume and Flavor Materials of Natural Origin (Elizabeth, N.J., 1960) and in "Flavor and Fragrance Materials--1991", Allured Publishing Co. Wheaton, Ill. USA.

Some non-limiting examples of useful volatile materials whose molecular weight is less than 175 are:

Material	Molecular Weight
ethyl acetate	88
iso-amyl alcohol	88
2-methylpyrazine	94
cis 3-hexenol	100
C6-aldehyde	100
C6 alcohol	102
ethyl propionate	102
benzaldehyde	106
benzyl alcohol	108
C7-aldehyde	114
methyl amyl ketone	114
iso amyl formate	116

ethyl butyrate	116
Indole	117
acetophenone	120
phenyl ethyl alcohol	122
styrallyl alcohol	122
Veltol TM	126
methyl hexyl ketone	128
3-methyl 3-methoxy butanol	128
ethyl amyl ketone	128
octenol JD	128
prenyl acetate	128
C8-aldehyde	128
amyl acetate	130
cinnamic aldehyde	132
phenyl propyl aldehyde	134
cinnamic alcohol	134
terpinolene	136
phenyl acetic acid	136
phenyl propyl alcohol	136
alpha pinene	136
benzyl formate	136
anisic aldehyde	136
d- limonene	136
Triplal TM	138
Cyclal C TM	138
Melonal TM	140
C-9 aldehyde	142
iso nonyl aldehyde	142
cyclo hexyl acetate	142
ethyl caproate	144
hexyl acetate	144
coumarin	146

methyl cinnamic aldehyde	146
cuminic aldehyde	148
benzyl acetone	148
geranyl nitrile	149
cuminy alcohol	150
benzyl acetate	150
Heliotropine TM	150
thymol	150
neral	152
synthetic vanillin	152
synthetic citral	152
rose oxide	154
geraniol	154
allyl caproate	156
Rosalva TM	156
tetrahydro myrcenol	158
yara yara	158
diethyl malonate	160
methyl cinnamate	162
Jasmorange TM	162
benzyl propionate	164
eugenol	164
ethyl vanillin	166
dihydrojasmone	166
geranic acid	168
methyl laltone	168
methyl nonyl ketone	170
methyl tuberate	170
hexyl butyrate	172
octyl-3-acetate	172
hydroxycitronellol	174
Fructose TM	174

Some non-limiting examples of useful materials that can be used that have a molecular weight higher than 175 are:

Material	Molecular Weight
benzal glyceryl acetal	180
anisyl acetate	180
terpinyl formate	182
geranyl formate	182
methyl diphenyl ether	184
delta undecalactone	184
allyl amyl glycolate	186
amyl caproate	186
Fraistone TM	188
Pelargene TM	188
Florhydral TM	190
ethyl hexyl ketone	190
ethyl phenyl glycidate	192
Verdyl acetate TM	192
dihydro beta ionone	194
iso-butyl salicylate	194
allyl cyclo hexyl propionate	196
myrcenyl acetate	196
citronellyl oxyacetaldehyde	198
citral dimethyl acetal	198
beta naphthyl iso butyl ether	200
tetrahydro linalyl acetate	200
amyl cinnamic aldehyde	202
Fruitaflor TM	202
Lilial TM	204
damascenone	204
methyl ionone	206

Cashmeran TM	206
Ebanol TM	206
phenoxy ethyl iso butyrate	208
iso amyl salicylate	208
Sandalore TM	210
propyl diantilis	210
benzyl benzoate	212
citronellyl propionate	212
myristic alcohol	214
Gelsone TM	214
hexyl cinnamic aldehyde	216
butyl butyryllactate	216
amyl cinnamate	218
hydroxycitronellal dimethyl acetal	218
beta methyl ional	220
Vetiverol TM	220
hexyl salicylate	222
geranyl crotonate	222
methyl jasmonate	224
linalyl butyrate	224
Hedione TM	226
Timberol TM	226
Floramat TM	228
benzyl salicylate	228
Fixal TM	230
Cetone V TM	232
cis carveol	232
Iso E Super TM	234
muscalone	234
geranyl tiglate	236
Cetalox TM	236
linalyl valerate	238

benzyl cinnamate	238
Thibetolide TM	240
phenyl ethyl phenylacetate	240
phenyl ethyl salicylate	242
Boisambrene TM	242
jasmonyl	244
Phantolid TM	244
methyl cedryl ketone	246
Aldrone TM	248
amyl cinnamic aldehyde dma	248
Dione TM	250
cedryl formate	250
ambrettolide	252
phenyl ethyl cinnamate	252
benzyl iso eugenol	254
hexadecanolide	254
Novalide TM	256
citronellyl ethoxalate	256
Fixolide TM	258
Galaxolide TM	258
rose acetate	262
ambrate	262
iso caryl acetate	264
cinnamyl cinnamate	264
ethyl undecylenate	266
Ethylene Brassylate TM	272
triethyl citrate	276
dihexyl fumarate	284
Okoumal TM	288
musk ketone	294
alpha Santalol TM	300
geranyl iso valerate	312

The solvent of the volatile liquid can be selected from many classes of volatile compounds that known to the art, for example, ethers; straight or branched chain alcohols and diols; volatile silicones; dipropylene glycol, triethyl citrate, ethanol, isopropanol, diethyleneglycol monoethyl ether, dipropylene glycol, diethyl phthalate, triethyl citrate, isopropyl myristate, etc.,

5 hydrocarbon solvents such as IsoparTM or other known solvents that have previously been used to dispense volatile actives from substrates. These solvents in general have a molecular weight between 20 and 400. They are selected specifically for each volatile liquid to achieve the performance and safety, (e.g. VOC and flash point) specified.

10 When the active is an insect repellent it can be composed of one or more compounds such as pyrethrum and pyrethroid type materials commonly now used in mosquito coils are likely to be the most useful for this purpose. Other insect control actives can be used, such as the repellents DEET, essential oils, such as citronella, lemon grass oil, lavender oil, cinnamon oil, neem oil, clove oil, sandalwood oil and geraniol.

15

When the active is an antimicrobial it can be composed of one or more of compounds such as essential oils such as rosemary, thyme, lavender, eugenic, geranium, tea tree, clove, lemon grass, peppermint, or their active components such as anethole, thymol, eucalyptol, farnesol, menthol, limonene, methyl salicylate, salicylic acid, terpineol, nerolidol, geraniol, and mixtures

20 thereof. benzyl alcohol, ethylene glycol phenyl ether, propylene glycol phenyl ether, propylene carbonate, phenoxyethanol, dimethyl malonate, dimethyl succinate, diethyl succinate, dibutyl succinate, dimethyl glutarate, diethyl glutarate, dibutyl glutarate, dimethyl adipate, diethyl adipate, dibutyl adipate, or mixtures thereof one or more aldehydes selected from cinnamic aldehyde, benzaldehyde, phenyl acetaldehyde, heptylaldehyde, octylaldehyde, decylaldehyde,

25 undecylaldehyde, undecylenic aldehyde, dodecylaldehyde, tridecylaldehyde, methylnonyl aldehyde, didecylaldehyde, anisaldehyde, citronellal, citronellyloxyaldehyde, cyclamen aldehyde, alpha-hexyl cinnamic aldehyde, hydroxycitronellal, alpha-methyl cinnamic aldehyde, methylnonyl acetaldehyde, propylphenyl aldehyde, citral, perilla aldehyde, tolylaldehyde, tolylacetaldehyde, cuminaldehyde, LilialTM, salicyl aldehyde, alpha-amylcinnamic aldehyde

30 and HeliotropineTM.

Other volatile actives can be used alone or in combination with the above actives, for example decongestants such as menthol, camphor, eucalyptus etc., malodor counteractants such as are

trimethyl hexanal, other alkyl aldehydes, benzaldehyde, and vanillin, esters of alpha-, beta-unsaturated monocarboxylic acids, alkyl cyclohexyl alkyl ketones, derivatives of acetic and propionic acids, 4-cyclohexyl-4-methyl-2-pentanone, aromatic unsaturated carboxylic esters, etc.

5

Care must be taken when designing the volatile liquid in that they pose no danger to the public. This is done by ensuring that the said volatile liquid has a flashpoint greater than about 60°C as determined by Test Method ASTM D93.

- 10 The transfer medium must have external capillary channels, that is, channels of capillary dimensions provided on an external surface of the medium such that a liquid will exhibit capillary flow within them. These may be provided by any suitable means, such as moulding and engraving. The transfer medium may be any suitable form of such medium, but is preferably one of two kinds:

15

1. The type in which a member bearing external capillary channels contacts directly a liquid in a reservoir, and the liquid rises in the capillary channels and evaporates into the atmosphere. An example of such a type is described in US 4,193,350
- 20 2. A type in which the liquid in the reservoir is taken therefrom by a porous wick in contact with it, there being mounted on the wick a capillary sheet whose external capillary channels are in liquid transfer contact with the wick, the liquid passing from the wick to the capillary channels and evaporating into the atmosphere. An example of such an apparatus is described in UK patent application GB 0306449

25

For the working of this invention, it is essential that the volatile liquid have a surface tension of 40 dynes/cm maximum and that the plastics material have a surface energy of 45 dynes/cm maximum. It has been found that this combination of parameters allows for an especially good dissemination of a liquid into an atmosphere. The invention therefore also provides a method of

30 disseminating a volatile liquid into an atmosphere by evaporation from a transfer member having surface capillary channels, the volatile liquid being such that at least 30% by weight of the materials comprising it have a molecular weight of 175 maximum, and that it has a surface

tension of less than 40 dynes/cm, and the transfer member being of plastics material having a surface energy of less than 45 dyne/cm.

The provision of a volatile liquid having the abovementioned characteristics is well within the skill of the art.

Preferably the liquid has a surface tension of less than 40 dyne/cm, and is more preferably within the range 20-35 dynes/cm. All surface tensions referred to herein are measured on a Fisher Surface Tensiomat model number 21 at 25°C.

10

It is further preferred that the volatile liquid have a viscosity of less than 10 centistokes per second at 25°C as measured on a Cannon-Fenske Viscometer according to Test Method ASTM D 445.

15 The plastics materials for use in this invention preferably have a surface energy of from 15-45 dyne/cm. The surface energy of a plastics material is dependent upon its molecular structure and is a measure of the ability of a surface to be wetted. The more inert is a plastics material chemically, the lower is its surface energy. Thus, materials such as polyethylene, polypropylene and PTFE have low surface energies, whereas the plastics with more polar groups have higher surface energies. Preferably the surface energy lies in the range of from 30-45 dynes/cm and more preferably from 30-35 dyne/cm. Some suitable materials for the purposes of this invention are shown in the following table:

Material Name	Example Material Trade Name(s)	Supplier	Surface Energy Dynes/cm
Polytetrafluoroethylene PTFE	TEFLON FEP106N	DU PONT	18
Polyethylene PE (HDPE)	BOREALIS MG 9641-R	NORTHERN PLASTICS	30
Polyethylene PE (LDPE)	IPETHENE 320	CARMEL OLEFINS	30
Polyethylene PE (LLDPE)	LL6201	EXXON MOBIL	30
Polystyrene PS	PS 146L	NOVA CHEMICALS	36

Polyvinylchloride PVC			41
Polyethylene terephthalate PET	RADITER	RADICI (PLASTRIBUTIO N)	42
Polycarbonate PC	LUPILON S- 3000R	MITSUBISHI POLYMERS	40
Polyvinylpropylene PP	EXP 058	EXXON MOBIL	32

(TEFLON, BOREALIS, IPETHENE, RADITER and LUPILON are trade marks)

Suitable transfer members may be easily fabricated by known means, for example, by the
5 methods described in the abovementioned US 4,913,350 and GB application 0306449.

The invention is further described by the following non-limiting examples.

Example 1.

10

Capillary sheets of polypropylene BP 400Ca 70, measuring 2.5 cm x 7.5 cm and having a
surface energy of 32 dyne/cm, were immersed to a depth of 1.25 cm. into 10g of a number of
vanilla fragrances containing different amounts of volatile materials with a MW less than 175.

The quantity of fragrance diffused into the air was determined by weighing the container with
15 fragrance and capillary. The following results were obtained after 4 days.

Fragrance	% MW < 175	Wt loss g/day
A1	14.5	0.35
A2	34.5	0.87
A3	53.6	0.64
A4	61.6	0.69
A5	69.05	1.10
A6	75.6	0.84
A7	81.6	0.86
A8	93.5	0.97
A9	93.5	1.07

This shows that, for effective transmission of fragrance into the atmosphere, the composition must have at least 30% of the fragrance materials with a molecular weight of less than 175.

Example 2

- 5 Two frusto-conical polyester wicks were placed in 11.5 g of A1 and A2 fragrances in BarexTM containers and allowed to equilibrate overnight. 1.5 mm thick polypropylene external capillary sheets with a central hole that allowed them to be fitted to the wicks were placed thereon, and the quantity of fragrance diffused per day was measured. The results after 6 days are shown below:

10

Fragrance	% MW < 175	Weight Loss g/day
A1	14.5	0.4
A2	35.5	1.0

For a hybrid system i.e. one in which the transport of the fragrance is via a porous wick and the diffusion is via an external capillary, good diffusion is obtained when the fragrance has a quantity of components with a MW < 175 is around 30% or higher

15

Example 3.

Capillary sheets of polypropylene BP 400Ca 70, measuring 2.5cm x 7.5 cm external capillary and having a surface energy of 32 dyne/cm, were immersed to a depth of 1.25cm into 10g of a
20 series of fragrances having more than 30% components with MW < 175, but with different surface tensions. The surface tension was measured at 25 C using a Fisher Surface Tensiomat model number 21.

The quantity of fragrance diffused into the air was determined by weighing the container with
25 fragrance and capillary. The following results were obtained after 2 days:

Fragrance	Wt Loss g/day	Surface tension Dynes/cm
B1	1.1	35.6
B2	0.7	38.2
B3	0.5	41.2
B4	0.5	42.2

This shows the advantage of having a surface tension below 40, and preferably below 38, dynes/cm.

5

Example 4.

A capillary sheet of polypropylene BP 400Ca 70, measuring 2.5cm x 7.5 cm and having a surface energy of 32 dyne/cm, was immersed to a depth of 1.25cm into 10g of a series of
10 fragrances having more than 30% components with MW < 175, but with different viscosities, The viscosity was measured using a Cannon-Fenske Viscometer by ASTM D 445 .

The quantity of fragrance diffused into the air was determined by weighing the container with fragrance and capillary. The following results were obtained after 2 days:

15

Fragrance	Wt Loss g/day	Viscosity Cs/s
C1	0.4	13.7
C2	0.4	11.9
C3	0.4	10.6
C4	0.9	8.2
C5	1.1	6.0

For good diffusion, the viscosity of the fragrance should be below 10 Cs/s.

Example 5.

Capillary sheets with different surface energies were set up as per example 1 with fragrance D (% Components MW<175 > 30, surface tension 37 dynes/cm and viscosity 5.7 Cs/s) and

5 fragrance E (% Components MW<175 > 30, Viscosity 2.9 cS/s and surface tension 34.5 dynes/sec), respectively. The fragrances had an oil-soluble dye added and the height to which it rose (as a percentage of the height of the capillary) after 6 minutes was measured and recorded, and is shown in the following tables.

10 Table 5 Effect of surface energy on diffusion of fragrance D

Plastic	Surface energy dyne/cm	Rise 6 min
PP BP 400	32	100(3)
PETG	41	81
PB ABS	46	59

The 100% rise in PP BP 400 was achieved after only 3 minutes.

Table 6 Effect of surface energy on diffusion of fragrance E.

Plastic	Surface energy dynes/cm	Rise 6 min
PP BP 400	32	100(1.2)
PETG	41	100(2)
PB ABS	46	41

15

100% rise was found after 1.2 min and 2 min, respectively for PP BP 400 and PETG.

This shows that the surface energy of the plastics material of the external capillary should be below 45 dynes/cm, preferably below 40 dynes/cm.

Claims:

1. An apparatus adapted to disseminate volatile liquid into an atmosphere from a reservoir,
the transfer to atmosphere being at least partially achieved by means of a transfer member
5 having external capillary channels, characterised in that
 - (a) at least 30% by weight of the materials comprising the volatile liquid have a molecular
weight of 175 maximum and the volatile liquid has a surface tension of less than 40
dynes/cm; and
 - 10 (b) the transfer member is of plastics material having a surface energy of less than 45
dyne/cm.
2. An apparatus according to claim 1, in which the surface tension of the liquid is from 20-
35 dynes/cm.
- 15 3. An apparatus according to claim 1, in which the surface energy of the plastics material is
from 15-45 dynes/cm.
4. An apparatus according to claim 3, in which the surface energy lies in the range of from
20 30-45 dynes/cm.
5. An apparatus according to claim 4, in which the surface energy lies in the range of from
30-35 dynes/cm.
- 25 6. An apparatus according to claim 1, in which the volatile liquid has a viscosity of less than
10 centistokes per second at 25°C.
7. An apparatus according to claim 1 in which the transfer member bears external capillary
channels, which directly contact a liquid in a reservoir, and the liquid rises in the capillary
30 channels and evaporates into the atmosphere.
8. An apparatus according to claim 1, in which the liquid in the reservoir is taken therefrom
by a porous wick in contact with it, there being mounted on the wick a capillary sheet

whose external capillary channels are in liquid transfer contact with the wick, the liquid passing from the wick to the capillary channels and evaporating into the atmosphere.

9. A method of disseminating a volatile liquid into an atmosphere by evaporation from a transfer member having surface capillary channels, the volatile liquid being such that at least 30% by weight of the materials comprising it have a molecular weight of 175 maximum, and that it has a surface tension of less than 40 dynes/cm, and the transfer member being of plastics material having a surface energy of less than 45 dyne/cm.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/CH2004/000647

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 A61L9/00 A61L9/12 A61L9/04 A01M1/20 A01M13/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A61L A01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 02/34302 A (JONES STUART MICHAEL RUAN ; RECKITT BENCKISER UK LTD (GB)) 2 May 2002 (2002-05-02)	1-6,8
Y	page 5, line 22 - page 6, line 10; claim 5; figures	9
X	GB 2 357 973 A (RECKITT BENCKISER ; RECKITT & COLMANN PROD LTD (GB)) 11 July 2001 (2001-07-11)	1-7
Y	page 1, lines 3-8 page 3, line 1 - page 4, line 9 page 7, line 24 - page 8, line 12; figures	9
X	WO 02/30220 A (WOLPERT CHRISTOPHER J ; DIAL CORP (US); TRIPLETT CARL (US); HE MENGTAO) 18 April 2002 (2002-04-18)	1-7
Y	figures; example 1	9
	-/--	

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

A document defining the general state of the art which is not considered to be of particular relevance

E earlier document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

G document member of the same patent family

Date of the actual completion of the international search

22 December 2004

Date of mailing of the international search report

13/01/2005

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Persichini, C

INTERNATIONAL SEARCH REPORT

International Application No
PCT/CH2004/000647

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 2 180 450 A (MEDICAL EXPRESS) 1 April 1987 (1987-04-01)	1-7
Y	page 3, lines 10-30 page 4, lines 10-15	9
Y	US 4 913 350 A (PURZYCKI KENNETH L) 3 April 1990 (1990-04-03) cited in the application figures; example 4	9

INTERNATIONAL SEARCH REPORT

 International Application No
 PCT/CH2004/000647

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 0234302	A	02-05-2002	AU 9577301 A	06-05-2002
			BR 0114841 A	16-09-2003
			CA 2426220 A1	02-05-2002
			EP 1328299 A1	23-07-2003
			WO 0234302 A1	02-05-2002
			GB 2370225 A , B	26-06-2002
			MX PA03003562 A	15-10-2003
			US 2004060997 A1	01-04-2004
			ZA 200303199 A	26-04-2004
GB 2357973	A	11-07-2001	NONE	
WO 0230220	A	18-04-2002	AU 1153002 A	22-04-2002
			AU 1775902 A	22-04-2002
			CA 2425304 A1	08-04-2003
			CA 2425589 A1	18-04-2002
			EP 1334313 A2	13-08-2003
			WO 0231413 A2	18-04-2002
			WO 0230220 A1	18-04-2002
			US 2002136886 A1	26-09-2002
			US 2002136542 A1	26-09-2002
GB 2180450	A	01-04-1987	AU 599928 B2	02-08-1990
			AU 6407686 A	07-04-1987
			EP 0278951 A1	24-08-1988
			WO 8701565 A1	26-03-1987
US 4913350	A	03-04-1990	AT 101799 T	15-03-1994
			AU 619530 B2	30-01-1992
			AU 3195589 A	05-10-1989
			CA 1316454 C	20-04-1993
			DE 68913294 D1	31-03-1994
			DE 68913294 T2	07-07-1994
			WO 8908462 A1	21-09-1989
			EP 0386158 A1	12-09-1990
			JP 2756328 B2	25-05-1998
			JP 2503522 T	25-10-1990